

Supplementary Table S1. Penalty points for screening of 92 illegal adulterants in soft-gel-type dietary supplements using QuEChERS, EMR-Lipid, and DLLME followed by UHPLC-Q/TOF-MS.

	Penalty points		
	QuEChERS	EMR-Lipid	DLLME
Reagents			
Acetonitrile	8	4	4
formic acid	1		
MgSO ₄	0		
NaCl	0		
PSA	2		
GCB	1		
C18	0		
EMR-Lipid material kit		0	
De-ionized water		0	
Chloroform			3
Instruments			
Energy			
Ultrasonic	0	0	0
Vortex	0	0	0
Centrifuge	2	2	2
LC-Q/TOF-MS	3	3	3
Occupation hazard	0	0	0
Waste			
1 - 10 mL (g)	3	3	3
Recycling	0	0	0
Total penalty points	25	17	20
Analytical Eco-scale total score	75	83	80

According to Gałuszka et al. (2012).

PSA, primary secondary amine; GCB, graphitized carbon black

Supplementary Table S2. Retention times, molecular formulae, and accurate mass measurements of 92 illegal adulterants obtained by UHPLC-Q/TOF-MS.

No.	Analyte	RT (min)	Molecular formula	Theoretical m/z ion (Da)	Error (ppm)
1	Metformin	1.24	C ₄ H ₁₁ N ₅	130.1087	3.23
2	Amiloride HCl	1.34	C ₆ H ₈ ClN ₇ O	230.0552	0.71
3	4-Dimethylaminoantipyrine	1.37	C ₁₃ H ₁₇ N ₃ O	232.1444	-0.27
4	Theophylline	1.38	C ₇ H ₈ N ₄ O ₂	181.0720	0.55
5	Ephedrine HCl	1.43	C ₁₀ H ₁₅ NO	166.1227	-2.75
6	Acetaminophen	1.54	C ₈ H ₉ NO ₂	152.0706	3.96
7	Triamterene	1.57	C ₁₂ H ₁₁ N ₇	254.1149	-2.01
8	Captopril	2.00	C ₉ H ₁₅ NO ₃ S	218.0846	2.42
9	Yohimbin	2.27	C ₂₁ H ₂₆ N ₂ O ₃	355.2016	2.04
10	Hydroxywardenafil	2.35	C ₂₃ H ₃₂ N ₆ O ₅ S	505.2228	1.33
11	Thioquinapiperifil	2.39	C ₂₄ H ₂₈ N ₆ OS	449.2118	1.56
12	Bambuterol	2.54	C ₁₈ H ₂₉ N ₃ O ₅	368.2180	0.58
13	Vardenafil	2.62	C ₂₃ H ₃₂ N ₆ O ₄ S	489.2279	0.12
14	Carbodenafil	2.71	C ₂₄ H ₃₂ N ₆ O ₃	453.2609	1.70
15	Brompheniramine	2.99	C ₁₆ H ₁₉ BrN ₂	319.0804	0.19
16	Bupropion HCl	3.08	C ₁₃ H ₁₈ ClNO	240.1150	-4.67
17	Triprolidine	3.16	C ₁₉ H ₂₂ N ₂	279.1856	3.49
18	Norneovardenafil	3.25	C ₁₈ H ₂₀ N ₄ O ₄	357.1577	2.32
19	Hongdenafil	3.43	C ₂₅ H ₃₄ N ₆ O ₃	467.2765	0.88
20	Dimethylacetildenafil	3.54	C ₂₅ H ₃₄ N ₆ O ₃	467.2765	1.35
21	Ketotifen fumarate salt	3.61	C ₁₉ H ₁₉ NOS	310.1260	1.62
22	Icariin	3.63	C ₃₃ H ₄₀ O ₁₅	677.2440	-0.41
23	Astemizole	3.73	C ₂₈ H ₃₁ FN ₄ O	459.2555	0.78
24	Propranolol	3.85	C ₁₆ H ₂₁ NO ₂	260.1645	0.45
25	Modafinil	3.86	C ₁₅ H ₁₅ NO ₂ S	296.0716	0.89
26	Oxohongdenafil	3.97	C ₂₅ H ₃₂ N ₆ O ₄	481.2558	-0.22
27	Olopatadine	4.00	C ₂₁ H ₂₃ NO ₃	338.1751	1.42
28	Sildenafil	4.31	C ₂₂ H ₃₀ N ₆ O ₄ S	475.2122	-1.09
29	Avanafil	4.42	C ₂₃ H ₂₆ ClN ₇ O ₃	484.1858	1.15
30	Dimethylsildenafil	4.51	C ₂₃ H ₃₂ N ₆ O ₄ S	489.2279	-0.08
31	Diphenhydramine	4.57	C ₁₇ H ₂₁ NO	256.1696	2.00
32	Methylprednisolone	4.90	C ₂₂ H ₃₀ O ₅	375.2166	1.35
33	Carbamazepine	4.90	C ₁₅ H ₁₂ N ₂ O	237.1023	-0.28
34	Dimenhydrinate	5.07	C ₁₇ H ₂₁ NO	256.1696	-2.05
35	Betamethasone	5.15	C ₂₂ H ₂₉ FO ₅	393.2072	-0.30
36	Eplerenone	5.16	C ₂₄ H ₃₀ O ₆	415.2115	1.23
37	Acetaminotadalafil	5.17	C ₂₃ H ₂₀ N ₄ O ₅	433.1506	0.79
38	Dexamethasone	5.25	C ₂₂ H ₂₉ FO ₅	393.2072	0.49
39	Udenafil	5.29	C ₂₅ H ₃₆ N ₆ O ₄ S	517.2592	-0.30
40	Promethazine	5.33	C ₁₇ H ₂₀ N ₂ S	285.1420	3.87
41	Demethyltadalafil	5.38	C ₂₁ H ₁₇ N ₃ O ₄	376.1292	4.48
42	Piroxicam	5.39	C ₁₅ H ₁₃ N ₃ O ₄ S	332.0700	3.16
43	Paroxetine	5.48	C ₁₉ H ₂₀ FN ₂ O ₃	330.1500	-2.15
44	Beclomethasone	5.50	C ₂₂ H ₂₉ ClO ₅	409.1776	-1.42
45	4-Isopropylantipyrine	5.60	C ₁₄ H ₁₈ N ₂ O	231.1492	0.84
46	Phenolphthalein	5.63	C ₂₀ H ₁₄ O ₄	319.0965	-0.69
47	Ketorolac	5.79	C ₁₅ H ₁₃ NO ₃	256.0968	1.21
48	Flunisolide	5.96	C ₂₄ H ₃₁ FO ₆	435.2177	1.50
49	Sulindac	5.97	C ₂₀ H ₁₇ FO ₃ S	357.0955	-0.80
50	Cyproheptadine	6.07	C ₂₁ H ₂₁ N	288.1747	1.33
51	Levothyroxine	6.07	C ₁₅ H ₁₁ I ₄ NO ₄	777.694	1.01
52	Bisacodyl	6.1	C ₂₂ H ₁₉ NO ₄	362.1387	-0.91
53	Boldenone	6.19	C ₁₉ H ₂₆ O ₂	287.2006	6.13
54	Desulfovardenafil	6.33	C ₁₇ H ₂₀ N ₄ O ₂	313.1659	2.27
55	Benzylsildenafil	6.44	C ₂₈ H ₃₄ N ₆ O ₄ S	551.2435	-0.38
56	Xanthoanthrafil	6.44	C ₁₉ H ₂₃ N ₃ O ₆	390.166	0.95
57	Didesmethylsibutramine	6.58	C ₁₅ H ₂₂ ClN	252.1514	-2.19
58	Prednisone-21-acetate	6.68	C ₂₃ H ₂₈ O ₆	401.1959	0.18
59	Fexofenadine	6.71	C ₃₂ H ₃₉ NO ₄	502.2952	-2.24
60	Fluoxetine HCl	6.72	C ₁₇ H ₁₈ F ₃ NO	310.1414	1.06

Supplementary Table S2. (continued)

No.	Analyte	RT (min)	Molecular formula	Theoretical m/z ion (Da)	Error (ppm)
61	Dapoxetine	6.77	C ₂₁ H ₂₃ NO	306.1852	0.15
62	Mirodenafil	6.77	C ₂₆ H ₃₇ N ₅ O ₅ S	532.2588	3.60
63	Prednisolone-21-acetate	6.77	C ₂₃ H ₃₀ O ₆	403.2115	2.50
64	Beclomethasone-21-hemisuccinate	6.78	C ₂₆ H ₃₃ ClO ₈	509.1937	3.11
65	Cortisone-21-acetate	6.78	C ₂₃ H ₃₀ O ₆	403.2115	0.30
66	Sibutramine	7.10	C ₁₇ H ₂₆ ClN	280.1827	-1.60
67	Sertraline HCl	7.10	C ₁₇ H ₁₇ Cl ₂ N	306.0811	-2.60
68	Homotadalafil	7.11	C ₂₃ H ₂₁ N ₃ O ₄	404.1605	2.00
69	Boldione	7.15	C ₁₉ H ₂₄ O ₂	285.1849	-4.17
70	Meloxicam	7.40	C ₁₄ H ₁₃ N ₃ O ₄ S ₂	352.0421	0.69
71	Mibolerone	7.90	C ₂₀ H ₃₀ O ₂	303.2319	0.55
72	Danazol (M)	8.00	C ₂₁ H ₂₈ O ₂	313.2162	0.01
73	Chlorosibutramine	8.24	C ₁₇ H ₂₅ Cl ₂ N	314.1437	1.55
74	Spironolactone	8.30	C ₂₄ H ₃₂ O ₄ S	341.2111	0.30
75	Fluocinonide	8.83	C ₂₆ H ₃₂ F ₂ O ₇	495.2189	-1.48
76	Calusterone	8.85	C ₂₁ H ₃₂ O ₂	317.2475	0.03
77	Clostebol	8.98	C ₁₉ H ₂₇ ClO ₂	323.1772	-1.16
78	Cyclopentyltadalafil	9.01	C ₂₆ H ₂₅ N ₃ O ₄	444.1918	1.79
79	Chloropretadalafil	9.28	C ₂₂ H ₁₉ ClN ₂ O ₅	427.1055	-3.71
80	Betamethasone-17-valerate	9.50	C ₂₇ H ₃₇ FO ₆	477.2647	-1.08
81	Diclofenac	9.51	C ₁₄ H ₁₁ Cl ₂ NO ₂	296.0240	1.60
82	Indomethacin	9.52	C ₁₉ H ₁₆ ClNO ₄	358.0841	3.54
83	Acceclofenac	9.67	C ₁₆ H ₁₃ Cl ₂ NO ₄	354.0295	-2.47
84	Imidazosagatriazinone	9.95	C ₁₇ H ₂₀ N ₄ O ₂	313.1659	0.31
85	Terfenadine	10.15	C ₃₂ H ₄₁ NO ₂	472.321	0.42
86	Phenylbutazone	10.16	C ₁₉ H ₂₀ N ₂ O ₂	309.1598	2.46
87	Norbolethone	10.24	C ₂₁ H ₃₂ O ₂	317.2475	0.38
88	Betamethasone-21-valerate	10.35	C ₂₇ H ₃₇ FO ₆	477.2647	0.18
89	Betamethasone dipropionate	10.54	C ₂₈ H ₃₇ FO ₇	505.2596	-0.60
90	Beclomethasone dipropionate	11.00	C ₂₈ H ₃₇ ClO ₇	521.2301	-1.79
91	Rimonabant	12.24	C ₂₁ H ₃₂ O ₂	463.0854	0.39
92	Testosterone-17-propionate	12.90	C ₂₂ H ₃₂ O ₃	345.2424	-1.70